

Effects of Substrate Interactions on In-plane and Out-of-plane Order in Thin Films of Lamellar Copolymers

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Thin films of lamellar copolymers can assemble into well-oriented perpendicular domains when confined between two "neutral" interfaces, and controlling this behavior is critical for applications in semiconductor lithography. We examined the ordering of poly(styrene-*b*-methyl methacrylate) (PS-PMMA) lamellar copolymers confined between a neutral free surface and a "nearly neutral" brushed poly(styrene-*r*-methyl methacrylate) silicon substrate. The PS-PMMA film thickness (t) and brush grafting density (Σ) were systematically varied to examine their impacts on in-plane and out-of-plane ordering. Samples were characterized with a combination of high resolution microscopy, x-ray reflectivity, and grazing-incidence small angle X-ray scattering (GISAXS). Perpendicular lamellae were detected at the neutral free surface of all samples, and there were no combinations of t and Σ that drove island or hole formation. In-plane order at the top of the film (quantified through calculation of orientational correlation lengths) improved with tn , where the exponent n increased from approximately 0.75 to 1 as Σ decreased from 0.6 to 0.2 nm². Out-of-plane defects such as bent or tilted domains were detected in all films through detailed analysis of GISAXS data. The width of the out-of-plane orientation distribution did not follow a simple scaling law with t or Σ , but the maximum extent of disorder was observed in samples where the underlying brushes had low Σ . These studies demonstrate that weakly preferential interactions at the substrate can contribute to both in-plane and out-of-plane disorder.